

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant: Seiki TAMURA et al. **Confirmation No:** 1093
Application No.: 10/598,108 **Group Art Unit:** 1796
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Title: Method Of Manufacturing Purified Products Of Liquid Medium-Chain Alkyl-Modified Polydimethylsiloxane And Cosmetics Prepared Therefrom

DECLARATION UNDER 37 CFR § 1.132

Mail Stop: Amendment
Commissioner for Patents
P.O. Box 1450
Alexandria, Virginia 22313-1450

Dear Sir:

I, Seiki Tamura, hereby state that:

1. I am a citizen of Japan.
2. I have a Bachelor of Engineering degree from Kyoto University of Japan. I am currently employed in a Polymer Development Specialist role for Dow Corning Toray Co., Ltd. of Tokyo, Japan. I have worked in the silicone field for 17 years and I have been employed by Dow Corning Toray for the past 6 years.
3. I am a named inventor of the pending U.S. Patent Application, Serial No. 10/598,108, and a person highly skilled in the siloxane art, including alkyl-modified siloxanes,

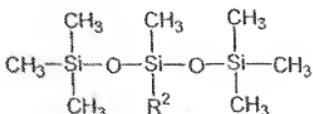
components thereof, processes for producing alkyl-modified siloxanes, and compositions including alkyl-modified siloxanes.

4. I am aware of, have read, and understand the disclosure of JP2003-048813 (the '813 publication), which is entitled "WATER-IN-OIL TYPE EMULSION COMPOSITION AND EMULSION COSMETIC".

5. I am aware of, have read, and understand the disclosure of JP2003-306550 (the '550 publication), which is entitled "METHOD FOR DEODORIZING ALKYL METHYL SILOXANE WAX".

6. In relevant part, our invention, which is a method of manufacturing a final product (having high purity) of a liquid medium-chain alkyl-modified polysiloxane, includes the step of synthesizing the liquid medium-chain alkyl-modified polysiloxane by carrying out a hydrosilylation reaction between 1,1,1,3,5,5-heptamethyltrisiloxane and an α -olefin having 4 to 12 carbon atoms to form a crude product. Our method further includes the step of hydrogenating the crude product, and optionally, stripping the crude product, to form the final, high purity, product. Hydrogenation of the crude product requires a catalyst, such as a Raney nickel catalyst.

7. The final product includes a polysiloxane of the general formula (3):



where R^2 represents an alkyl group with 4 to 12 carbon atoms (imparted by the corresponding α -olefin). After formation, the final product is generally free of a specific, unpleasant, odor/scent and is essentially odorless altogether. The same is generally true when the final product is stored over a period of time, or a subsequent product including the final product, such as a cosmetic composition, is stored over a period of time. One of the sources of the unpleasant odor in the crude product is residual α -olefin left over from the hydrosilylation reaction which has subsequently oxidized over time. Typically, our final product includes ≥ 98 parts by weight of the polysiloxane (3), based on 100 parts by weight of the final product. Said another way, our final product typically has a purity of 98% or greater, which is considered "high purity".

8. For the reasons described in Paragraphs 9 through 19 immediately below, our invention is unique and distinguishable from the composition of the '813 publication and from the method of the '550 publication.

9. First, while the '550 publication generally teaches steps of hydrogenation and stripping to deodorize alkyl methyl siloxane waxes, it was not previously known, at the time of our invention, that a similar method could be used to deodorize alkyl methyl siloxane liquids. Said another way, our method was not previously known or even contemplated for use with alkyl methyl siloxane liquids. Specifically, a method such as ours was not even sought after because conventional methods of deodorizing siloxane liquids were already known and utilized in the art.

10. Prior to our method, the primary solution for addressing the odor problem of siloxane liquids, if such a problem was present at all, was to simply apply heat and vacuum to the crude product to remove the excess α -olefin and/or oxidized α -olefin, as well as any other causative reactants or byproducts. In addition or as an alternate to applying heat and vacuum, masking agents could also be used to cover the odor, such as a perfume. An additional option was to simply control the molar ratio of reactants such that there is only a very small amount of excess α -olefin utilized in forming the crude product.

11. After reviewing the '813 publication and the '550 publication, I can find no teaching or even a suggestion in one of these documents that would lead one skilled in the art, such as myself, to refer to the other document. In the '813 publication, there is no teaching or even a suggestion of how to form its silicone oils, such as which raw materials are used, what amounts of raw materials are used, etc. As such, it is not clear whether the initial silicone oils or the subsequent composition of the '813 publication has a problematic odor, or any odor at all for that matter. For example, it is completely plausible that the silicone oils are essentially odor free based on method of manufacture, e.g. using equal or slightly excess molar parts of an α -olefin relative to a Si-H reactant. Alternatively, it can be inferred that the silicone oils of the '813 publication are put into subsequent compositions in such a time frame that does not even allow for one to appreciate odor or odor development, i.e., a time frame shorter than one necessary for the odor to reach detectable levels. As such, when referring to the '813

publication, one of skill in the art, such as myself, would not have turned to the teachings of the '550 publication regarding deodorization when no odor was to be deodorized.

12. The same is true in the opposite reference scenario. For example, in the '550 publication, it is made explicit that the method taught therein is for polysiloxane waxes, and the '550 publication even describes the differences between waxes and oils/liquids (see, e.g. paragraph [0002] of the '550 publication). Further, the '550 publication focuses on having longer-chain alkyl groups of 14 to 50 carbons relative to most preferred shorter alkyl groups of the '813 publication having 8 to 12 carbon atoms. Polysiloxane waxes and polysiloxane liquids are not equivalents, in both the chemical and physical sense. As such, when referring to the '550 publication, one of skill in the art would not have turned to the teachings of '813 publication when the '813 publication is concerned with siloxane liquids and immediate emulsions thereof rather than concerned with siloxane waxes, which are drastically different in both chemical and physical properties.

13. Further yet, referring to the working examples of the '550 publication, the '550 publication relies on Dow Corning Toray siloxane wax, specifically "AMS-C30 Cosmetic Wax". Referring to the Material Safety Data Sheet (MSDS) and information sheet of this product, it is described that AMS-C30 has a "characteristic odor", and is a "mixture of methyl siloxane and organic compound". In addition, physically, AMS-C30 is described as a "waxy flake". This odor description makes sense because AMS-C30 includes 40 to 70 wt % of C24-54 α -olefin, remainder wt % alkyl siloxane wax. As such, one skilled in the art appreciates that

the method of the '550 publication is directed toward siloxane waxes of very low purity having a very large amount of residual long-chain α -olefin, which inherently have strong odors. Not only do these waxes have the odor because of the presence of such olefins, but physically, because these waxes are in fact "waxy", such olefins are physically bound up until released, such as when using the waxes in manufacturing, where the waxes are melted thereby releasing the previously entrapped odors.

14. In contrast, siloxane liquids can have far less residual α -olefin, if any at all, as introduced above. For example, in Practical Example 1 of our pending application, we utilized 1000 parts of 1,1,1,3,5,5-heptamethyltrisiloxane and 555 parts of 1-octene. Theoretically, the reaction product of these two reactants has no more than ~10 wt % residual 1-octene, assuming none of the 1-octene flashes off during storage. Therefore, AMS-C30 has at least 4 to 7 times more residual α -olefin than the crude product of our Practical Example 1. That being said, it can be inferred that the residual 1-octene in Practical Example 1 would not leave the crude product during such a time frame. Obviously, if an odor is present, so must be a volatized chemical compound imparting that odor. This time frame could be very long in duration depending on a variety of variables, to the point that one would not have necessarily recognized that our crude product even developed an odor over time. In addition, these liquids physically cannot bind odors in the same manner as waxes. Said another way, waxes and liquids have completely different release profiles in terms of volatile components, e.g. components that cause an odor such as raw α -olefins.

15. For the reasons described in Paragraphs 9 through 14 above, one skilled in the art, prior to our invention, would not have applied the method of the '550 publication to the siloxane oils of the '813 publication. In addition, even if one were to combine the teachings of the '550 publication and the '813 publication, the results would not have been predictable as described in Paragraphs 16 and 19 immediately below.

16. First, any alteration to a chemical, let alone a chemical reaction, entails some degree of unpredictability. This is especially the case when one refers to siloxanes in general, with near infinite species of siloxanes and combinations of such siloxanes in the art each having, and imparting, specific and different physical and chemical properties. An easy example is illustrated above, where it is shown that while polysiloxanes may have similar structures, they can be of completely different physical state, e.g. a wax vs. a liquid. Obviously, when using each of these polysiloxanes in a reaction, they will behave differently, based on chemical structure and physical state, which will impact reaction kinetics. For example, the wax has a different chemical structure than that of the liquid, and heat must be applied to the wax to reach the same physical state of the liquid. Both of these differences will impact the subsequent reaction of the siloxane wax differently relative to subsequent reaction of the siloxane liquid.

17. Surprisingly, in our invention, I, along with my co-inventor, discovered at least the absence of an expected property. Specifically, as detailed in the data presented below, we discovered that α -olefins with 4 to 12 carbon atoms could easily be removed from the crude product of our invention, but that higher α -olefins, such as those with 16 carbon atoms could

not be easily removed from the crude product of our invention. For example, one of skill in the art might predict that α -olefins having 20 or more carbon atoms would be difficult to remove from crude products after an addition reaction based on higher molecular weights relative to α -olefins having less than 20 carbon atoms. Even for one of skill in the art, however, it would be difficult to predict that they could not remove substantially all α -olefins with 13 to 19 carbon atoms from the same system by a stripping step and that they could easily obtain a final product with very high purity by removing α -olefins with 4 to 12 carbon atoms from the crude product. For example, based on the teachings of the '550 publication, one skilled in the art would have predicted that even when the reaction products of the '550 publication included remaining α -olefins, such α -olefins could be removed by some aggressive stripping step unless the α -olefins had 20 or more carbon atoms in a long-chain.

18. Tables I and II below illustrate additional examples to emphasize the different results of our process depending on the type of α -olefin employed. Specifically, Table I illustrates two inventive examples each employing a 12 carbon α -olefin (1-dodecene), and Table II illustrates a comparative example employing a 16 carbon α -olefin (1-hexadecene). The inventive examples are prepared according to the method of our invention, and similar to the examples described in the specification of our pending application. Inventive Example 2 is merely a scaled up version of Inventive Example 1. Comparative Example 3 is of the same scale as Inventive Example 1. Each of the three examples included a hydrogenation reaction that was carried out for 6 hours at 140 °C.

TABLE I

MD ^{R*1} M (R*1 = -C ₁₂ H ₂₅)	Initial Odor Level	Purity of Product (GC %)	Residual C ₁₂ Hydrocarbon (GC %)	Conditions for Stripping
Ex. 1-1 Reaction Mixture	Very bad odor	82.3	17.7	—
Ex. 1-1 Crude Product	Odor	99.3	0.6	140 °C, 3 hr 1 mm Hg
Ex. 1-2 Final Product (High Purity)	Odorless	99.3	0.6	115 °C, 2 hr 40 mm Hg
Ex. 2-1 Reaction Mixture	Bad odor	96.5	3.5	—
Ex. 2-1 Crude Product	Odor	98.3	1.6	140 °C, 3 hr 10 mm Hg
Ex. 2-2 Final Product (High Purity)	Odorless	99.5	0.2	150 °C, 2 hr 10 mm Hg

TABLE II

MD ^{R*2} M (R*2 = -C ₁₆ H ₃₃)	Initial Odor Level	Purity of Product (GC %)	Residual C ₁₆ Hydrocarbon (GC %)	Conditions for Stripping
Ex. 3-1 Reaction Mixture	Bad odor	89.7	10.0	—
Ex. 3-1 Crude Product	Odor	90.0	9.8	160 °C, 8 hr 1 mm Hg
Ex. 3-2 Final Product (Low Purity)	Odorless	90.2	9.6	165 °C, 4 hr 1 mm Hg

19. As shown in Tables I and II above, removability of residual hydrocarbons (α -olefin or its corresponding alkane) having 12 carbon atoms is very different from removability of residual hydrocarbons having 16 carbon atoms. For example, highly purified final products

(~99%) were obtained in Inventive Examples 1 and 2, but not in Comparative Example 3 (~90%). The final products of Inventive Examples 1 and 2 include heptamethyl dodecyl trisiloxane, and the final product of Comparative Example 3 includes heptamethyl hexadecyl trisiloxane. While all of the final products were initially odorless after production, surprisingly, Comparative Example 3-2 still included almost 10% residual C16 hydrocarbons which subsequently would cause an unpleasant odor over time. This was surprising because the residual hydrocarbon content remained high even after employing much more aggressive stripping conditions to both the crude and final products formed from the C16 α -olefin relative to the stripping conditions employed on the crude and final products formed from the C12 α -olefin. As described above, one of skill in the art would have predicted that aggressive stripping would have easily increased the purity of the final product of Example 3; however, this is not the case.

20. Conclusions

For the reasons described in Paragraphs 9 through 19 above, our invention is unique and distinguishable from the '813 publication and the '550 publication. Specifically, neither the '813 publication nor the '550 publication teaches or suggests that hydrogenation and stripping can be applied to siloxane liquids formed from the reaction of 1,1,1,3,5,5,5-heptamethyltrisiloxane and an α -olefin having 4 to 12 carbon atoms. In addition, I, along with my co-inventor, first discovered that not all α -olefins having less than 20 carbon atoms can be easily removed from the crude product such that our invention presents new and

unexpected results relative to the prior art including both the '813 publication and the '550 publication.

21. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information are believed to be true, and further that these statements were made with the knowledge that willful and false statements and the like are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or patent issued thereon.

Respectfully submitted,

Dated November 25, 2010

Seiki Tamura

Seiki Tamura